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Performance Characterization of a Water-based Multiplicity Counter

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1. ABSTRACT

In recent years the severe shortage of ^{3}He has necessitated the fast development of new neutron detection technologies. Here we report the performance characteristics of a water-based neutron detecting multiplicity counter for the non-destructive assay of fissile sources. We measured an absolute neutron detection efficiency of 28% and ^{60}Co rejection/suppression factor of ~10^{8} to 1. For sources with higher background gamma ray intensities, the neutron efficiency was 22% ± 1% up to a ^{60}Co equivalent rate of 4 MBq. We also present the sensitivity to small spontaneous fission sources such as ^{252}Cf and ^{240}Pu. By simulating a selection of isotopes and activities, we determine whether they can be distinguished by the neutron multiplicity distribution. If source related gamma ray rates are low, we have found that the multiplicity distributions of ^{240}Pu, ^{252}Cf and background should be distinguishable for source fission rates as low as ~10 Hz, corresponding to approximately 24 milligrams of ^{240}Pu.

2. INTRODUCTION

Coincidence counting of neutron pairs is an effective way to non-destructively determine the amount of fissile material within a sample of special nuclear material (SNM) [1]. Multiplicity counting is more versatile and precise, but also more demanding, requiring the detection of three or more neutrons per single fission event. Detecting *n* correlated neutrons from a single fission depends on the *n*th power of the detection efficiency, which is the critical determining factor in evaluating the utility of a particular neutron multiplicity detection technique.

In recent years the severe shortage of ^{3}He has been a great concern for governments and organizations involved in nuclear security. ([2],[3],[4]). ^{3}He detectors are uniquely suited for neutron detection, since they are insensitive to gamma rays, have a high neutron capture cross section, and are safe and non cryogenic. In particular, tightly packed arrays of ^{3}He tubes, surrounded by moderating material, are highly efficient. They have been in wide use since the 1970s to measure neutron multiplicities from fission chains, and hence the fissile content of both fresh and spent nuclear fuel, as well as other fissile material matrices. ^{3}He and polyethylene based well counting systems range in efficiency from 10% to 50%, depending on how tightly the tubes are packed and the gas density. Highly efficient and large systems, however, require the use of a large fraction of the yearly supply of ^{3}He and have become prohibitively expensive. In recent years the number of competing neutron detection techniques has proliferated in response to the ^{3}He shortage. Most are not ready for widespread use. Boron based systems such as BF_3 and ^{10}B tubes/planes are either toxic or relatively inefficient. Scintillator-based solutions generally rely on differences in signal pulse shape to discriminate against gamma rays, placing severe limits on the event rate that can be tolerated before pileup issues dominate. Germanium or silicon based detectors are small, reducing their overall efficiency. Given that the ^{3}He shortage is projected to continue for the

foreseeable future, alternative techniques are clearly needed, and are being actively pursued by various end users.

Recently we characterized the performance of a water Cherenkov-based neutron detector as a multiplicity well counter [5]. We reported that for sources that produce a low rate of gamma ray emission, the absolute neutron detection efficiency was 28%. The gamma ray rejection factor, a common metric for comparison of ^{3}He alternatives, was 10^{8}:1 for ^{60}Co. Both numbers are competitive with current ^{3}He-based systems and certainly compare favorably with non-^{3}He-based systems. For high activity sources, such as spent fuel, consistent detection efficiencies of 22% ± 1% were observed for ^{60}Co equivalent rates below ~ 4 MBq.

In this paper we use an event timing simulation that accurately reproduces the inter-event time distribution of a ^{252}Cf source to predict the multiplicity distribution we expect to observe from ^{240}Pu. The multiplicity distributions derived from the simulation are then used to determine the sensitivity of the detector, in terms of ^{240}Pu mass. We believe the results offer a uniquely beneficial solution to the ^{3}He problem, and fully justify further development of the technology.

3. THE DETECTOR

In order to perform this study, we reconfigured an existing water-based antineutrino detector at LLNL, tailoring its design to suit multiplicity-counting scenarios. The detector is comprised of 1.02 m^3 pure DI water doped with 0.5% gadolinium-chloride ($GdCl_3$) contained within a stainless steel tank (121.9 cm x 91.4 cm x 119.4 cm). The complete design was described in detail in [5]. A sealed polypropylene source deployment well (walls 1.2 cm thick) extends 73 cm down into the tank from the top, allowing the deployment of dry sources 19 cm in diameter to the center of the detector. If neutrons are emitted from a source in the well, they are moderated in the polypropylene and then the water, then captured on dissolved gadolinium. The resulting ~ 8 MeV gamma ray shower produces Cherenkov light which is detected by the PMTs. Figure 1 shows a schematic and picture of the detector prior to filling.

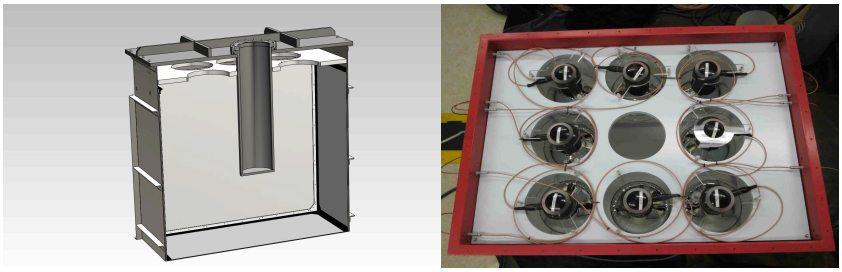

Figure 1: A schematic (left) of the detector showing a cut away of the 73 cm deep source deployment well/cavity and PMT placement (PMTs not shown). To the right is the finished detector immediately after PMT placement inside and prior to the installation of the lid and well.

4. CHARACTERISTIC RESPONSE TO NEUTRONS AND GAMMA RAYS

A 1.0 μCi ^{252}Cf source producing 4400 neutrons per second was used to determine the detector response to neutron captures. The energy deposited by a neutron capture was measured in terms of

the number of detected photoelectrons, and shown in Figure 2, together with a measured ^{60}Co spectrum. The neutron capture spectrum extends up to ~200 photoelectrons. The ^{60}Co spectrum, however, only extends as far as 50 photoelectrons. The no-source background signal, which is always present, results primarily from cosmic ray particles such as muons, neutrons, and high-energy gamma rays incident on the detector. After applying a neutron selection cut (between 50 and 200 photoelectrons), the no-source background rate is 155 Hz ± 0.2 Hz. The background rate and spectrum was remarkably stable over a long period. No measurable change was observed in almost three months, implying the background can be measured extremely accurately and subtracted from the source data.

A Geant4 simulation ([6], [7], [8]) of the detector was also constructed. Figure 3 shows a comparison between the simulated and measured detector spectrum from a neutron source placed in the base of the well. The simulation reproduces the spectrum very accurately above ~20 photoelectrons, an indication that the trigger reaches 100% efficiency at that level of signal. Because the simulation tracks all neutrons emitted from the source (including those that escape the detector entirely from the top of the well), the spectra do not match below 20 photoelectrons. The absolute neutron efficiency (28.0%) was calculated by counting the fraction of simulated neutrons that produce a response in the range 50 to 200 photoelectrons. It was also independently calculated from the nominal ^{252}Cf source intensity (28.5%). Both estimates included ALL (fission) neutrons emitted by the source, not simply the neutrons that hit the detector face (i.e. we are calculating the absolute, not intrinsic efficiency). We assumed the alpha-n source emission was negligible.

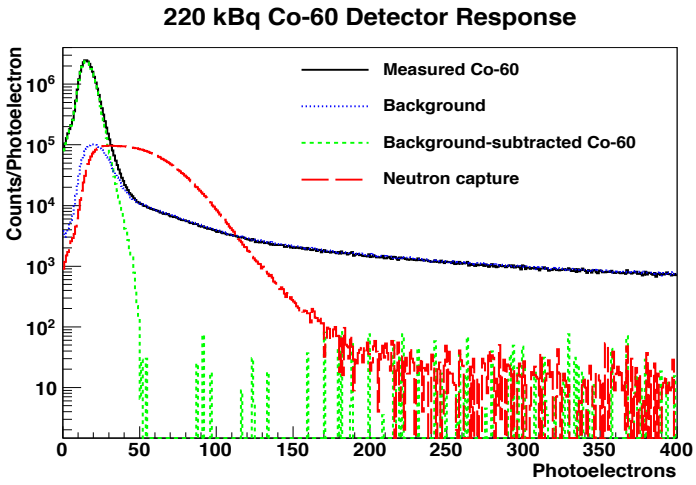

Figure 2: The detector spectral response of a 220 kBq ^{60}Co source for a one-hour data acquisition. Also shown is a one-hour background run, and the background subtracted ^{60}Co response. Shown for comparison is the neutron capture spectral response [5].

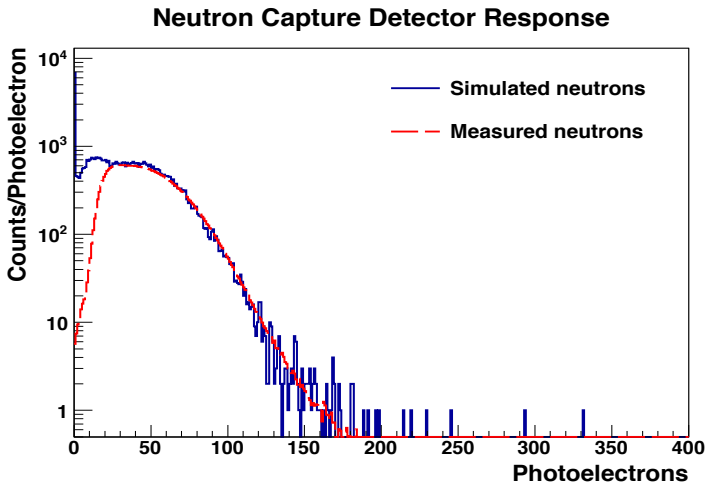

Figure 3: A comparison of the simulated (Geant4) and real data neutron capture detector spectrum.

5. TIMING MODEL AND NEUTRON MULTIPLICITY

Figure 4 shows the detected neutron multiplicity obtained with the ^{252}Cf source. The no source background is also included for comparison. A simple event timing simulation was employed to model the measured detector timing distribution, and to confirm if the multiplicity distribution could be reproduced. A published ^{252}Cf neutron multiplicity distribution [9] was used as input to the model (see Table 1). Our first task was to tune the model to reproduce the timing distribution obtained from the ^{252}Cf calibration data.

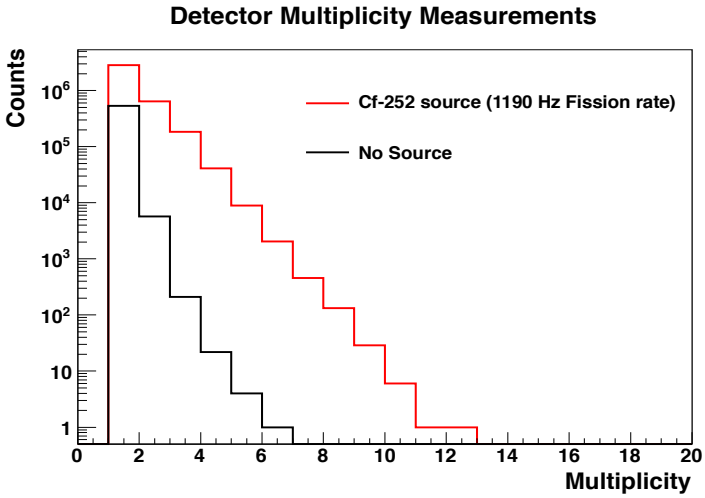

Figure 4: The neutron multiplicity distribution obtained from one-hour data acquisitions with and without a 1.0 µCi ^{252}Cf fission source in the source well. Based on calibrations of the source done in October 2007, the estimated source fission rate was 1190 spontaneous fissions per second.

Table 1: The fission neutron multiplicity probability distributions for ^{252}Cf and ^{240}Pu (taken from [9]).

P(n) Probability of multiplicity n	^{252}Cf	^{240}Pu
P(0)	0.002	0.066
P(1)	0.026	0.232
P(2)	0.127	0.329
P(3)	0.273	0.251
P(4)	0.304	0.102
P(5)	0.185	0.018
P(6)	0.066	0.002
P(7)	0.015	0
P(8)	0.002	0

The fixed and tuned inputs to our simulation are given in Table 1 and 2 respectively. Event times were sampled in the following way. Fission event times were sampled from a single exponential according to the nominal fission rate (1190 Hz, Table 2). The neutron multiplicity was sampled from the appropriate distribution in Table 1. After applying the measured detector efficiency (28%), surviving neutrons were assigned a neutron capture time (following the fission), sampled from the three component exponential of Equation 1. The parameters of this equation were then tuned with the aim of closely reproducing the observed ^{252}Cf detector inter-event time distribution of Figure 6:

Equation 1:

$$\textbf{\textit{Neutron Capture Time Following a Fission}} = \boldsymbol{A_1}(\boldsymbol{e}^{-t/\lambda_{Gd}}) + \boldsymbol{A_2}(\boldsymbol{e}^{-t/\lambda_H}) - \boldsymbol{A_3}(\boldsymbol{e}^{-t/\lambda_T})$$

Where $\boldsymbol{\lambda_{Gd}}$ is the mean thermal neutron capture time in a homogeneous water medium doped with 0.25% gadolinium and $\boldsymbol{t}$ the time since fission (both in μ-seconds), $\boldsymbol{\lambda_H}$ is the mean thermal capture time in polypropylene and $\boldsymbol{\lambda_T}$ the neutron thermalization time. The first exponential describes the timing of neutron captures on gadolinium, the second applies to the small subset of neutrons that thermalize in the polypropylene walls of the detector well, before drifting into the water and capturing on gadolinium. The subtracted third exponential approximates the neutron thermalization time in water. The ratios of the three amplitudes (A_1 = 1.0, A_2 = 0.015 and A_3 = 1.3) were also optimized from the tuning process and are presented in Table 2. The uncorrelated background (from detector measurements - 155 Hz) was sampled from an additional single exponential. Table 2 shows both the tuned (in blue) and fixed parameters (black). Figure 5 shows the functional form of our neutron capture time distribution.

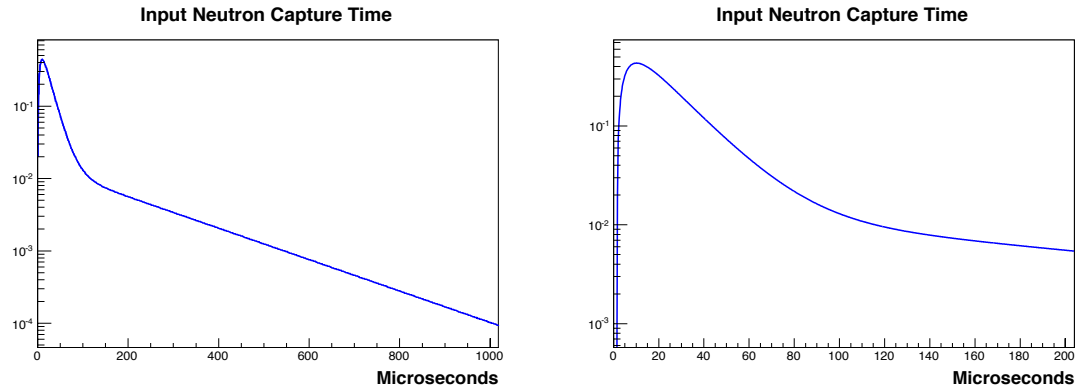

Figure 5: The functional form of Equation 1 shown over 1000 µ-seconds (left) and 200 µ-seconds (right). Three exponentials contribute to its shape - 1) the thermal neutron capture time in water doped with 0.25% gadolinium, 2) the thermal neutron capture time in polypropylene, and 3) the thermalization time of a fission neutron at 1 to 2 MeV (~5 µ-seconds).

Table 2: A list of the input parameters for our event timing model/simulation. The parameters shown in black were obtained from detector measurements, blue parameters were the optimized values obtained after allowing them to float over a small range during simulation tuning.

Model Input Parameter	**Input Value in Model**
^{252}Cf fission rate	1190 Hz
Assumed source multiplicity distribution	See Table 1 Multiplicity distributions
Uncorrelated background rate	155 Hz
Absolute neutron detection efficiency	28%
λ_{Gd}	18 µs
λ_{H}	200 µs
λ_{T}	5 µs
Relative exponential Amplitudes (A_1, A_2 and A_3)	1.0, 0.015, 1.3

Figure 6 shows the inter-event timing distribution obtained from our model compared to the measured ^{252}Cf distribution. Note that the detector inter-event time is used as our observable (for tuning purposes), not the time-since-fission. This is because we do not have a tag for the fission event. The real data has a cutoff at 3 µ-seconds due detector dead time following each event

imposed by the analysis. The simulation reproduces the observed distribution very well. Figure 7 shows a comparison of the simulated and detected multiplicity distributions. Multiplicities one, two and three are reproduced quite well. Higher multiplicities fall off in the simulation somewhat faster than real data. The reason for this is not understood at this time. However, we should note that the higher multiplicities are more likely to progressively magnify small detector effects that are not included in our simple model. Potential contributors include PMT after pulsing, or small uncertainties in the published multiplicity data in Table 1. Fortunately, only the lower order multiplicities are needed for a fissile material measurement.

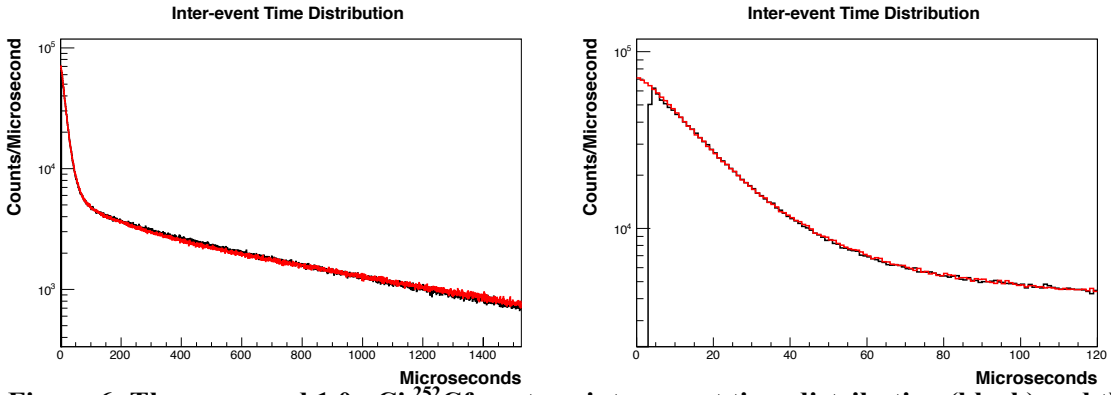

Figure 6: The measured 1.0 µCi ^{252}Cf neutron inter-event time distribution (black) and the inter-event time that results from the simulation after tuning (red). Note the 3 µ-second dead time in the real data distribution. This is included in our simulation as a post facto selection cut when determining the multiplicities.

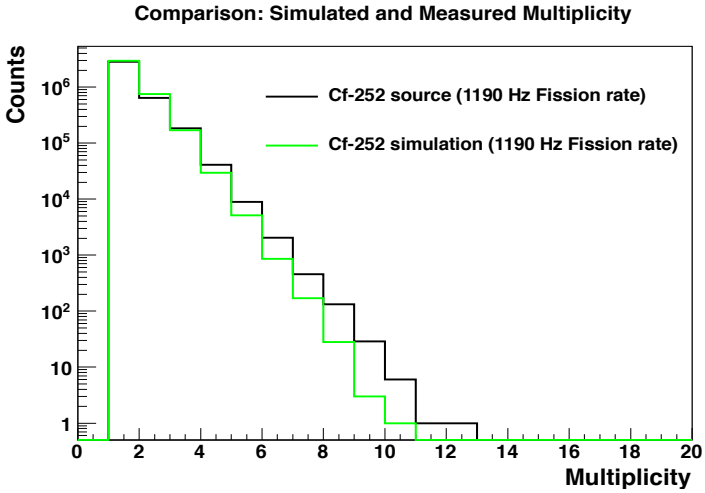

Figure 7: A comparison of the simulated (green) and measured (black) multiplicity distributions after tuning from a 1.0 µCi ^{252}Cf source, producing 1190 spontaneous fissions per second (see text for discussion).

Armed with our tuned simulation we can predict the multiplicities we expect to see from other spontaneous fission sources, such as ^{240}Pu. Figure 8 shows the multiplicities predicted by the model (singles, doubles and triples) for a variety of ^{240}Pu source intensities. The input multiplicities were again obtained from [9], and listed in Table 1 above. There are significant differences between the

^{252}Cf and ^{240}Pu input multiplicities. Our simulation indicates the detector may be very sensitive to these differences, even down to a fission rate of ~10 Hz, which is easily observable above the no source background. A fission rate of 10 Hz corresponds to approximately 24 milligrams of ^{240}Pu. Note that (statistical) error bars have been included in Figure 6, and are mostly small relative to the line thickness, only two are visible in the figure.

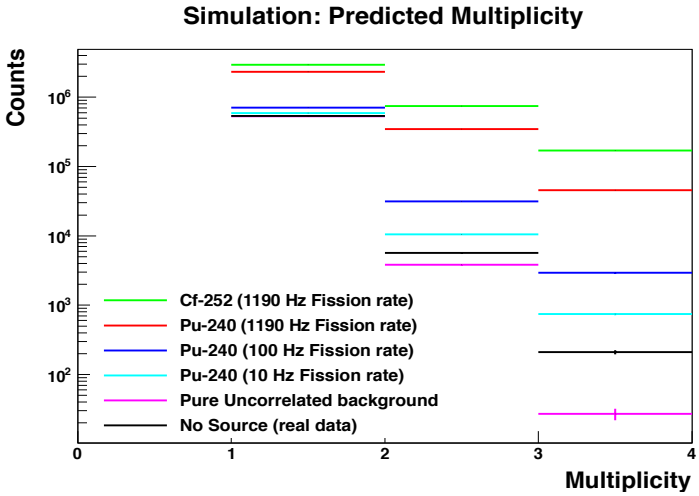

Figure 8: The predicted single, double and triple multiplicities obtained from our simulation of various ^{252}Cf and ^{240}Pu source intensities. The simulated source exposure time is equivalent to a one-hour data acquisition.

6. CONCLUSIONS

In conclusion, we report that under low background rate conditions, such as for a fresh fuel or low level waste samples, the absolute neutron detection efficiency of the water based well counter is 28%. At this efficiency the ^{60}Co gamma ray rejection factor is 10^8 to 1. Both numbers are competitive with current ^{3}He-based systems and certainly compare favorably with any non-^{3}He-based systems. We have simulated the performance of the LLNL water-based multiplicity detector by sampling a neutron capture time distribution comprised of a simple three component exponential function, and additionally sampling uncorrelated background from a single exponential function. The result accurately reproduces the observed inter-event time and multiplicity distributions of our ^{252}Cf detector data. We have used this tuned simulation to predict the multiplicities we expect to observe from a variety of ^{240}Pu source intensities. We conclude that the multiplicity distributions from this detector are likely to be highly sensitive to small quantities of ^{240}Pu, and also sensitive to small differences in fission source isotope. The multiplicity distributions indicate detector sensitivity should easily reach approximately 24 milligrams of ^{240}Pu after one-hour. Since the use of real world spent fuel sources was outside the scope of this work, further study is needed to determine how closely measurements from plutonium and MOX samples reproduce the predictions of our model.

In the near future our Geant4 simulation will be tested using the source term from a spent fuel source pin. The aim is to determine how much continuous background Cherenkov light will be produced in the detector as a result of a “hot” spent fuel source. We also hope to test the detector

with a small real world spent fuel source. In the future, it may be possible to extend the background range over which water-Cherenkov based systems may be applicable by taking advantage of segmentation (reducing pileup within each segment), using lead shielding in the source well, increasing the detector energy resolution using water soluble wavelength shifter, high quantum efficiency PMTs or more reflective materials.

7. ACKNOWLEDGEMENTS

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